LEAD SOIL TREND ANALYSIS
THROUGH MAY, 2006
EVALUATION BY INDIVIDUAL QUADRANT
Herculaneum Lead Smelter Site
Herculaneum, Missouri

Tetra Tech EM Inc. (Tetra Tech) was tasked by the U.S. Environmental Protection Agency (EPA) Region 7 Enforcement/Fund Lead Removal program to conduct a trend analysis of soil lead concentrations at selected locations within Herculaneum, Missouri (City). Specifically, the Tetra Tech Superfund Technical Assessment and Response Team (START) 3 was requested to review and analyze data that would enable EPA to determine if soil lead concentrations were increasing over time at a variety of locations within the City. Two tasks were identified: 1) perform a trend analysis for individual quadrants within each yard using the most current sampling data, and 2) estimate the range of monthly increase in lead concentrations for properties grouped into three categories based on distance from the smelter (less than or equal to 0.25 mile, 0.25 to 0.50 miles, and 0.50 to 0.75 miles). The assessment was conducted under the authority of the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 and the Superfund Amendments and Reauthorization Act of 1986. The project was assigned under START Contract No. EP-06-01, Task Order No. 0021.

Tetra Tech focused its analysis on one data set called "Recontamination." This data set includes results from a number of residential properties. The data were collected from four different quadrants at each property, and additional data for several properties came from samples collected in driveway areas outside the quadrants. Lead sampling was conducted at each location at varying intervals from the time removal activities were completed in early 2002 (sampling round 6). Sampling was conducted monthly prior to 2003, quarterly from 2003 to 2004, and semi-annually after October 2005 (sampling round 22). This report includes results for sampling conducted between August 2002 (sampling round 7) and May 2006 (sampling round 23). Due to the sequence of removal activities, not all properties underwent the same number of sampling events; the number of events ranged from 4 to 17 events per quadrant for individual properties. At many locations, some intervals within the series were omitted because of weather or access restrictions. The lead concentrations were determined by use of a portable X-ray fluorescence (XRF) instrument. Samples were collected and analyzed in accordance with the quality assurance project plan (QAPP) dated September 11, 2001.

This document presents the methods used to evaluate changes in soil lead concentrations following the removal activities, and the results of this analysis.

Methods

Trend tests were conducted for each property using data collected from round 7 (August 2002) through round 23 (May 2006). The non-parametric Mann-Kendall test was used to evaluate temporal trends for each sampled quadrant at the individual properties. The Mann-Kendall test is a widely used statistical test for detecting monotonic trends (that is, trends that are either increasing or decreasing) in time-series of data (Gilbert 1987; Helsel and Hirsch 1992; Gibbons 1994). Because the Mann-Kendall test uses only the relative magnitude of the data rather than their measured values, it has a number of desirable properties: the data need not be normally distributed; and the test is not significantly affected by outliers, missing data, or censored data. Censored data are treated in the Mann-Kendall test by setting all non-detect values to a concentration slightly below the minimum detected concentration. It should be noted that a minimum of four sampling events are required to perform this test, so properties with fewer than four rounds of sampling were not evaluated. Properties which were not sampled during round 23 were also excluded from the trend analysis.

For all properties where at least one quadrant showed a significant increasing trend based on the Mann-Kendall test, regression analysis was performed to estimate the monthly increase in lead concentration. This analysis was performed to provide rough estimates of the range of potential increase in lead concentrations for properties grouped according to distance from the smelter. Three distance categories were evaluated: less than or equal to 0.25 miles, 0.25 to 0.50 miles, and 0.50 to 0.75 miles. Because the purpose of this analysis was to only provide rough estimates of the rate of change in lead concentration, regression was performed on the data in original units (i.e., untransformed data). It should be noted that certain evaluation methods and diagnostic tools that are commonly used in linear regression analysis (e.g., evaluation of different transformations of the data, verification of model assumptions, and evaluation of outliers) were not used in this analysis.

For quadrants with detected data only, ordinary least squares (OLS) linear regression analysis was used. For quadrants with one or more censored (nondetect or ND) measurements, a censored maximum likelihood estimation (MLE) approach was used, following Helsel (2005). Censored MLE methods are increasingly being used in environmental assessment work, given the increased speed of modern personal computers and the enhanced capabilities that have been added into many commercial statistical software packages. As described in Helsel (2005), MLE regression techniques can be implemented using commercial software with capabilities for performing parametric survival analysis on interval-censored data. It should be noted that MLE regression for left-censored data is also referred to as "Tobit analysis" in the technical literature. MLE methods recognize each censored datum as an interval, bounded by zero at the lower limit and the detection or reporting limit at the upper limit. Application of OLS regression with censored data is contraindicated, as it requires substitution of an assumed value (typically zero, the detection limit, or one half the detection limit) for each censored datum, resulting in biased estimates for the regression parameters.

Results

Temporal trends in lead concentrations for 17 properties are summarized in Table 1 and Figure 1. The trend analysis identified 14 out of 17 properties where at least one quadrant showed a statistically significant increasing trend. No statistically significant decreasing trends were identified for any properties. Seven properties had increasing lead concentrations in all four quadrants: house numbers 5, 9, 18, 19, 20, 22, and 24. Two properties had increasing lead concentrations in three of four quadrants: house numbers 6 and 16. Four properties had increasing lead concentrations in two of four quadrants: house numbers 3, 7, 76 (only two quadrants evaluated), and 101. House number 15 had only one quadrant with an increasing trend in lead concentration. Three properties, house numbers 102, 103, and 104, showed no statistically significant trend in lead concentrations in any quadrant. All trend results are depicted graphically in Figure 1. Open symbols are used in Figure 1 to represent censored (nondetect) data, and solid symbols represent detected data.

Trend results reported for soil lead concentrations through sampling round 23 were similar to those reported during the last quarterly period, with the following exceptions. A single quadrant from each of four properties that did not show a significant trend in lead concentration from rounds 7 through 22, now show a statistically significant increase in lead concentration with the addition of the data from round 23. The properties include house numbers 6 (quadrant 4), 15 (quadrant 4), 24 (quadrant 1), and 101 (quadrant 3). Quadrant 4 from house number 102 showed a significant increasing trend in lead concentration from rounds 7 through 22, but this trend is no longer significant with the addition of data from round 23. Two additional properties, house numbers 103 and 104, now have 4 rounds of sampling and are being evaluated for the first time using the Mann-Kendall trend test. No significant increase in lead concentration was seen for any of the quadrants for these two properties.

The results of OLS and MLE regression analysis performed on properties that showed a significant increasing trend in lead concentration in at least one quadrant are provided in Table 2. The slope, intercept, standard error of the slope, and two-sided 95 percent confidence intervals for the slope estimates were calculated for 41 quadrants within 13 properties. Ranges for the monthly rates of increase in lead were 1.11 to 8.25 milligrams (mg)/month, 1.25 to 4.71 mg/month, and 0.78 to 7.80 mg/month, respectively, for properties located less than or equal to 0.25 miles, 0.25 to 0.50 miles, and 0.50 to 0.75 miles from the smelter. The upper 95 percent confidence limit (UCL) for the monthly rate of increase was also evaluated to estimate maximum potential rates of increase. Because of the variability in the individual estimates, the 50th, 75th, and 90th percentiles of the distribution of the individual UCLs within each distance category are also reported in Table 2. The 75th and 90th (in parentheses) percentile values for the monthly rate of increase for the properties grouped according to increasing distance from the smelter are 6.85 (10.85), 5.35 (6.19), and 3.88 (12.25) mg/month. It should be cautioned that these are considered rough estimates only, as no attempt was made to evaluate the validity of the regression model assumptions, or the uncertainty associated with the predicted rates of increase.

References:

Gibbons, R. D. 1994. *Statistical Methods for Groundwater Monitoring*. John Wiley & Sons, Inc. New York, New York.

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Helsel, D. 2005. *Nondetects and Data Analysis: Statistics for Censored Environmental Data*. John Wiley & Sons, Inc., New York, NY. 250 p.

Helsel, D. R. and R. M. Hirsh. 1992. *Statistical Methods in Water Resources*. Elsevier. New York, New York.

TABLE 1

RESULTS FO STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATON
INDIVIDUAL QUADARNS FOR SAMPLING ROUNDS 7 THROUGH 23
HERCULANEUM LEAD SMELTER SITE - HERCULANEUM, MISSOURI

Distance From Smelter (miles)[1]	House Number	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event		Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
					First	Last				
0.10	76	Q1	10	10	10/30/2003	05/18/2006	29	0.005	Yes	Increasing
		Q2	10	10	10/30/2003	05/18/2006	25	0.014	Yes	Increasing
0.20	20	Q1	16	16	08/26/2002	05/01/2006	74	0.001	Yes	Increasing
		Q2	16	16	08/26/2002	05/01/2006	72	0.001	Yes	Increasing
		Q3	16	16	08/26/2002	05/01/2006	84	<0.001	Yes	Increasing
		Q4	16	16	08/26/2002	05/01/2006	82	<0.001	Yes	Increasing
	101	Q1	9	9	12/22/2003	05/02/2006	12	0.130	No	N/A
		Q2	9	8	12/22/2003	05/02/2006	16	0.060	No	N/A
		Q3	9	9	12/22/2003	05/02/2006	20	0.022	Yes	Increasing
		Q4	9	9	12/22/2003	05/02/2006	22	0.012	Yes	Increasing
	102	Q1	9	9	12/22/2003	05/02/2006	14	0.090	No	N/A
		Q2	9	9	12/22/2003	05/02/2006	-8	0.238	No	N/A
		Q3	9	9	12/22/2003	05/02/2006	14	0.090	No	N/A
		Q4	9	9	12/22/2003	05/02/2006	12	0.130	No	N/A
0.25	5	Q1	16	13	08/26/2002	05/02/2006	88	<0.001	Yes	Increasing
		Q2	16	15	08/26/2002	05/02/2006	92	<0.001	Yes	Increasing
		Q3	16	16	08/26/2002	05/02/2006	85	<0.001	Yes	Increasing
		Q4	16	16	08/26/2002	05/02/2006	74	0.001	Yes	Increasing
	6	Q1	16	16	08/23/2002	05/02/2006	42	0.036	Yes	Increasing
		Q2	16	16	08/23/2002	05/02/2006	72	0.001	Yes	Increasing
		Q3	16	16	08/23/2002	05/02/2006	15	0.163	No	N/A
		Q4	16	16	08/23/2002	05/02/2006	46	0.026	Yes	Increasing
	22	Q1	15	15	08/26/2002	05/02/2006	51	0.009	Yes	Increasing
		Q2	15	15	08/26/2002	05/02/2006	53	0.007	Yes	Increasing
		Q3	15	15	08/26/2002	05/02/2006	58	0.004	Yes	Increasing
		Q4	15	15	08/26/2002	05/02/2006	57	0.004	Yes	Increasing
	24	Q1	13	13	11/07/2002	05/02/2006	30	0.042	Yes	Increasing
		Q2	13	13	11/07/2002	05/02/2006	56	0.001	Yes	Increasing
		Q3	13	13	11/07/2002	05/02/2006	40	0.012	Yes	Increasing
		Q4	13	12	11/07/2002	05/02/2006	43	0.007	Yes	Increasing
0.50	15	Q1	6	5	09/16/2002	05/02/2006	7	0.136	No	N/A
		Q2	6	6	09/16/2002	05/02/2006	8	0.102	No	N/A
		Q3	6	5	09/16/2002	05/02/2006	6	0.186	No	N/A
		Q4	6	5	09/16/2002	05/02/2006	11	0.028	Yes	Increasing
	16	Q1	14	10	09/16/2002	05/01/2006	27	0.071	No	N/A
		Q2	14	8	09/16/2002	05/01/2006	63	<0.001	Yes	Increasing
		Q3	14	8	09/16/2002	05/01/2006	44	0.010	Yes	Increasing
		Q4	14	10	09/16/2002	05/01/2006	59	0.001	Yes	Increasing
	19	Q1	16	15	08/22/2002	05/01/2006	51	0.016	Yes	Increasing
		Q2	16	13	08/22/2002	05/01/2006	53	0.013	Yes	Increasing
		Q3	16	13	08/22/2002	05/01/2006	51	0.016	Yes	Increasing
		Q4	16	15	08/22/2002	05/01/2006	66	0.003	Yes	Increasing

TABLE 1

RESULTS FO STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATON INDIVIDUAL QUADARNS FOR SAMPLING ROUNDS 7 THROUGH 23 HERCULANEUM LEAD SMELTER SITE - HERCULANEUM, MISSOURI

Distance From Smelter (miles)[1]	House Number	Quadrant	Number of Sampling Events[2]	Number of Detected Samples	Sampling Event		Mann-Kendall Test Statistic[3] (S)	Probability > S	Trend Significant?[4] (Yes/No)	Direction of Trend
					First	Last				
0.54	9	Q1	16	16	08/22/2002	05/01/2006	61	0.005	Yes	Increasing
		Q2	16	16	08/22/2002	05/01/2006	65	0.003	Yes	Increasing
		Q3	16	16	08/22/2002	05/01/2006	64	0.004	Yes	Increasing
		Q4	16	15	08/22/2002	05/01/2006	67	0.002	Yes	Increasing
0.60	18	Q1	17	17	08/23/2002	05/02/2006	56	0.015	Yes	Increasing
		Q2	17	16	08/23/2002	05/02/2006	47	0.033	Yes	Increasing
		Q3	17	17	08/23/2002	05/02/2006	65	0.006	Yes	Increasing
		Q4	17	17	08/23/2002	05/02/2006	72	0.003	Yes	Increasing
0.75	3	Q1	17	14	08/23/2002	05/02/2006	15	0.169	No	N/A
		Q2	17	15	08/23/2002	05/02/2006	66	0.005	Yes	Increasing
		Q3	17	16	08/23/2002	05/02/2006	33	0.084	No	N/A
		Q4	17	16	08/23/2002	05/02/2006	81	0.001	Yes	Increasing
0.79	103	Q1	4	1	03/28/2005	05/02/2006	3	0.271	No	N/A
		Q2	4	1	03/28/2005	05/02/2006	-1	0.500	No	N/A
		Q3	4	1	03/28/2005	05/02/2006	1	0.500	No	N/A
		Q4	4	2	03/28/2005	05/02/2006	3	0.271	No	N/A
0.80	7	Q1	17	17	08/23/2002	05/02/2006	21	0.142	No	N/A
		Q2	17	15	08/23/2002	05/02/2006	71	0.003	Yes	Increasing
		Q3	17	13	08/23/2002	05/02/2006	40	0.054	No	N/A
		Q4	17	12	08/23/2002	05/02/2006	60	0.010	Yes	Increasing
1.00	104	Q1	4	2	03/28/2005	05/01/2006	-3	0.271	No	N/A
		Q2	4	2	03/28/2005	05/01/2006	0	0.625	No	N/A
		Q4	4	1	03/28/2005	05/01/2006	-3	0.271	No	N/A

Notes:

1 Properties are ordered as a function of increasing distance from the smelter.
2 Trend tests were not conducted for properties with fewer than four rounds of sampling, or for properties not sampled during round 23.
3All censored (nondetect) measurements were set equal to a concentration slightly lower than the minimum detected value.
4 Monotonic trends are significant for probabilities less than or equal to 0.05; significant negative values for the Mann-Kendall test statistic indicate that trends are decreasing; and significant positive values for the Mann-Kendall test statistic indicate that trends are increasing.
NA No significant trend identified.

TABLE 2

RESULTS OF LINEAR REGRESSION ANALYSIS FOR ALL QUADRANTS SHOWING A SIGNIFICANT INCREASING MANN-KENDALL TREND TEST RESULT

Distance From Smelter (miles)	House Number	Quadrant	Number of Sampling Events	Regression Coefficients for Days Versus Concentration			Monthly Increase (mg/kg-month)	95 Percent Confidence Limits for Monthly Increase in Lead Concentrations		Percentiles for the Distribution of Estimated UCLs within Each Distance Group		
				Intercept	Slope	S.E. (Slope)		LCL	UCL	50	75	90
Less than or Equal to 0.25	76	Q1	10	45.78	0.16	0.05	4.68	1.24	8.12	5.28	6.85	10.85
	76	Q2	10	68.18	0.13	0.10	3.82	-2.94	10.59			
	20	Q1	16	94.89	0.13	0.03	3.99	1.90	6.09			
	20	Q2	16	55.84	0.27	0.05	8.01	4.73	11.29			
	20	Q3	16	111.47	0.17	0.04	5.12	2.76	7.48			
	20	Q4	16	82.13	0.28	0.04	8.25	5.48	11.02			
	101	Q3	9	11.92	0.12	0.04	3.46	0.94	5.97			
	101	Q4	9	-4.14	0.13	0.04	3.97	1.18	6.75			
	5	Q1	16	30.34	0.11	0.02	3.41	2.26	4.57			
	5	Q2	16	30.45	0.12	0.02	3.64	2.53	4.75			
	5	Q3	16	65.90	0.11	0.02	3.30	2.00	4.61			
	5	Q4	16	67.62	0.16	0.03	4.75	2.65	6.85			
	6	Q1	16	124.37	0.04	0.04	1.24	-1.24	3.73			
	6	Q2	16	83.85	0.11	0.03	3.41	1.54	5.28			
	6	Q4	16	80.12	0.04	0.02	1.11	-0.30	2.53			
	22	Q1	15	85.76	0.10	0.03	2.96	1.24	4.68			
	22	Q2	15	180.78	0.12	0.03	3.55	1.43	5.66			
	22	Q3	15	73.28	0.08	0.03	2.52	0.89	4.16			
	22	Q4	15	72.14	0.09	0.03	2.77	0.94	4.60			
	24	Q1	13	135.26	0.05	0.03	1.58	-0.29	3.44			
	24	Q2	13	27.08	0.14	0.03	4.30	2.61	5.99			
	24	Q3	13	64.01	0.04	0.01	1.14	0.43	1.84			
	24	Q4	13	60.04	0.07	0.02	1.97	0.53	3.42			
0.25 to 0.50	15	Q4	6	53.44	0.05	0.01	1.42	0.89	1.96	2.65	5.35	6.19
	16	Q2	14	28.52	0.16	0.02	4.71	3.33	6.09			
	16	Q3	14	61.74	0.04	0.02	1.33	0.29	2.36			
	16	Q4	14	58.91	0.14	0.03	4.24	2.30	6.19			
	19	Q1	16	55.13	0.04	0.01	1.28	0.44	2.13			
	19	Q2	16	41.67	0.06	0.01	1.94	1.04	2.84			
	19	Q3	16	40.91	0.04	0.02	1.25	0.04	2.46			
	19	Q4	16	55.63	0.07	0.02	2.04	0.94	3.13			
0.50 to 0.75	9	Q1	16	69.44	0.04	0.01	1.18	0.22	2.13	2.41	3.88	12.25
	9	Q2	16	64.23	0.08	0.02	2.48	1.23	3.73			
	9	Q3	16	93.17	0.26	0.08	7.80	2.48	13.13			
	9	Q4	16	86.85	0.10	0.02	2.97	1.63	4.31			
	18	Q1	17	73.60	0.05	0.02	1.40	0.31	2.49			
	18	Q2	17	52.84	0.05	0.02	1.57	0.54	2.60			
	18	Q3	17	71.84	0.03	0.01	0.78	0.18	1.37			
	18	Q4	17	59.49	0.05	0.01	1.62	0.90	2.34			
	3	Q2	17	51.56	0.04	0.01	1.34	0.41	2.27			
	3	Q4	17	45.22	0.04	0.01	1.32	0.71	1.92			

Notes:

- LCL Lower confidence limit
- MLE Maximum likelihood estimation
- ND Nondetect
- OLS Ordinary least squares
- S.E. Standard error of estimate
- UCL Upper confidence limit

OLS regression was used for cases where all results were detected. Censored MLE regression was used in all cases where one or more measurements were reported as below the detection limit (that is, "ND") following Helsel (2005). All analyses were performed on the data in original units.

Helsel, D. 2005. Nondetects and Data Analysis: Statistics for Censored Environmental Data. John Wiley & Sons, Inc., New York, NY. 250 pages.

FIGURE 1. Lead Concentration Trends From Round 7 Through 23

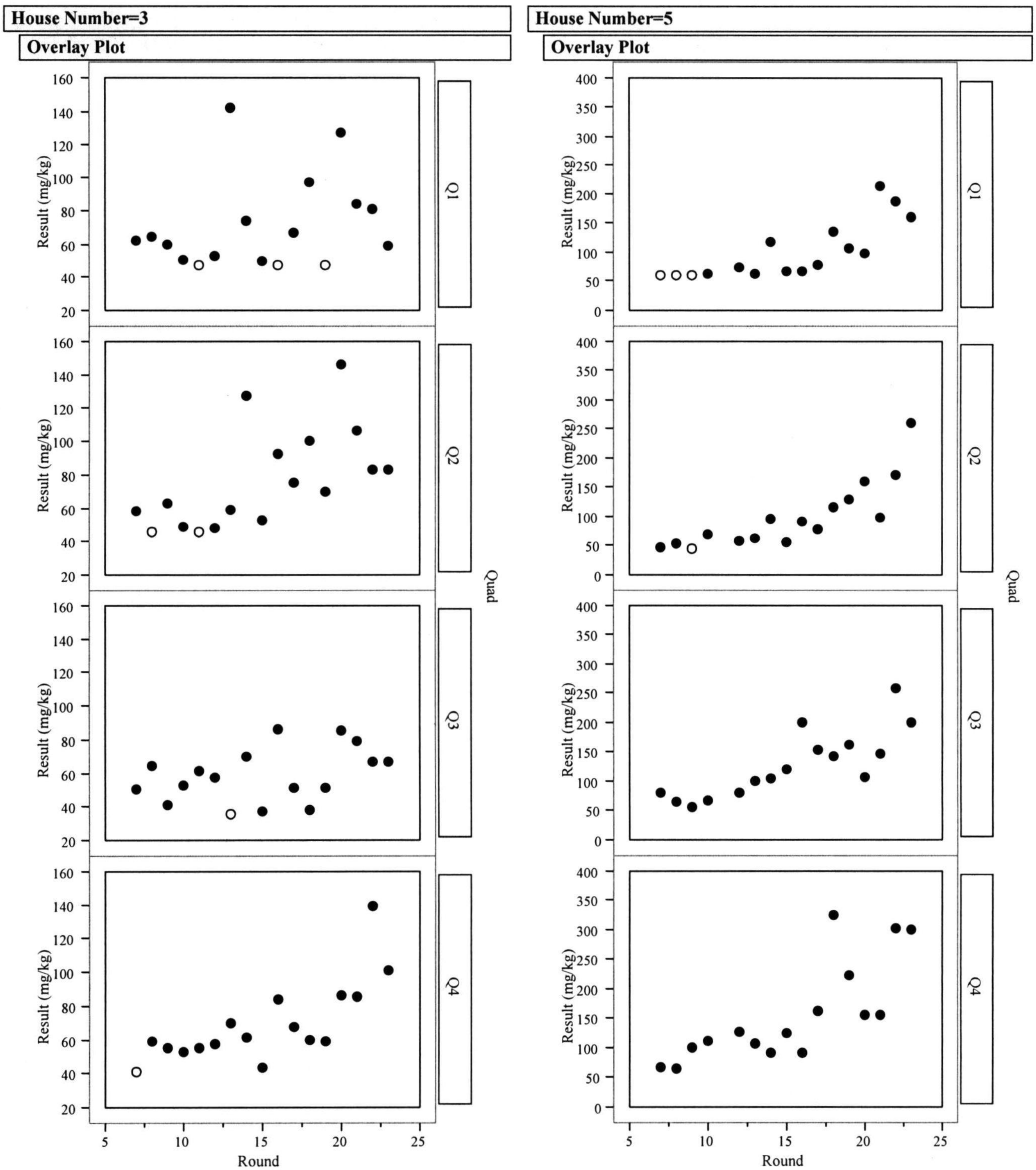


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)

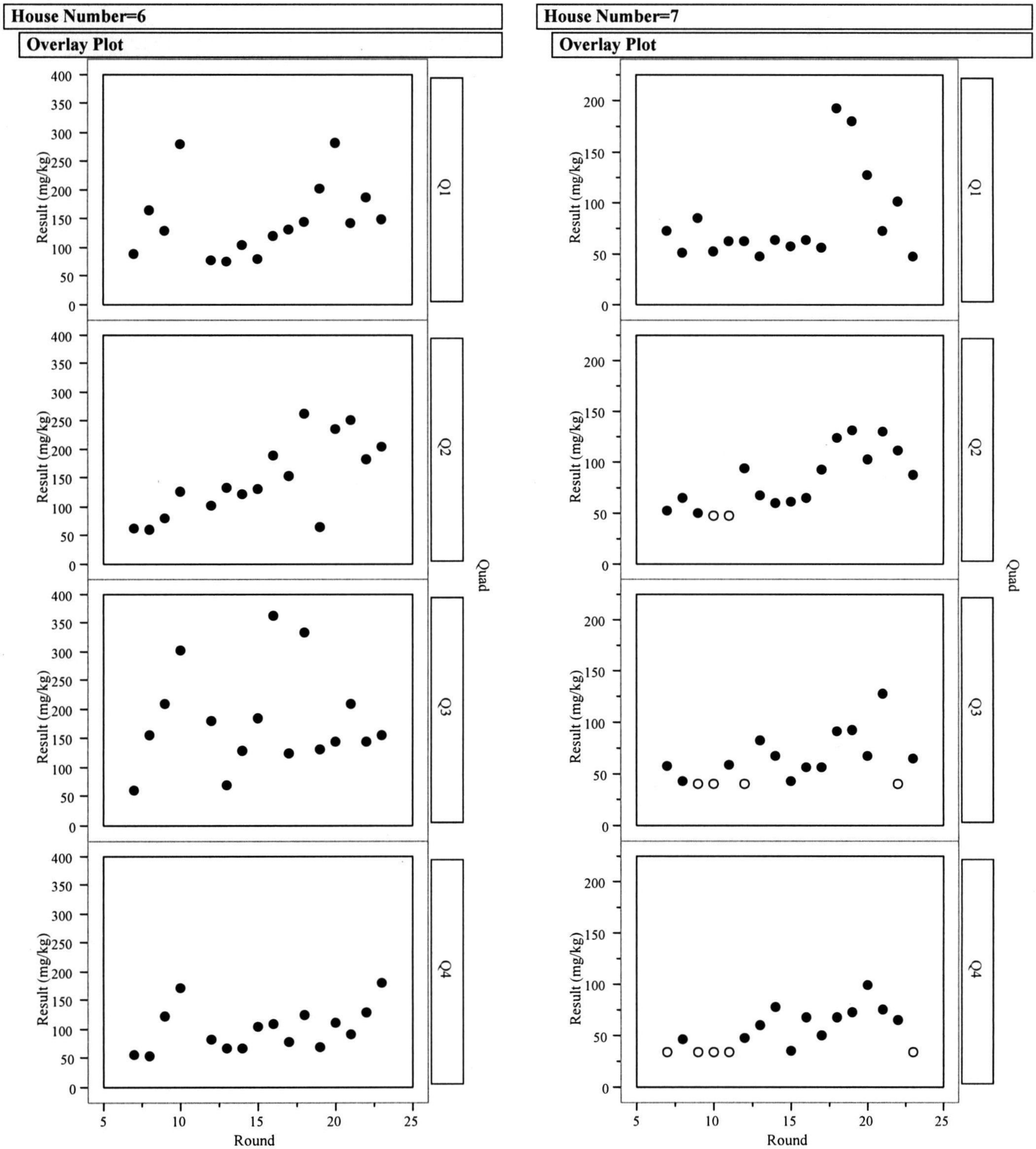


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)

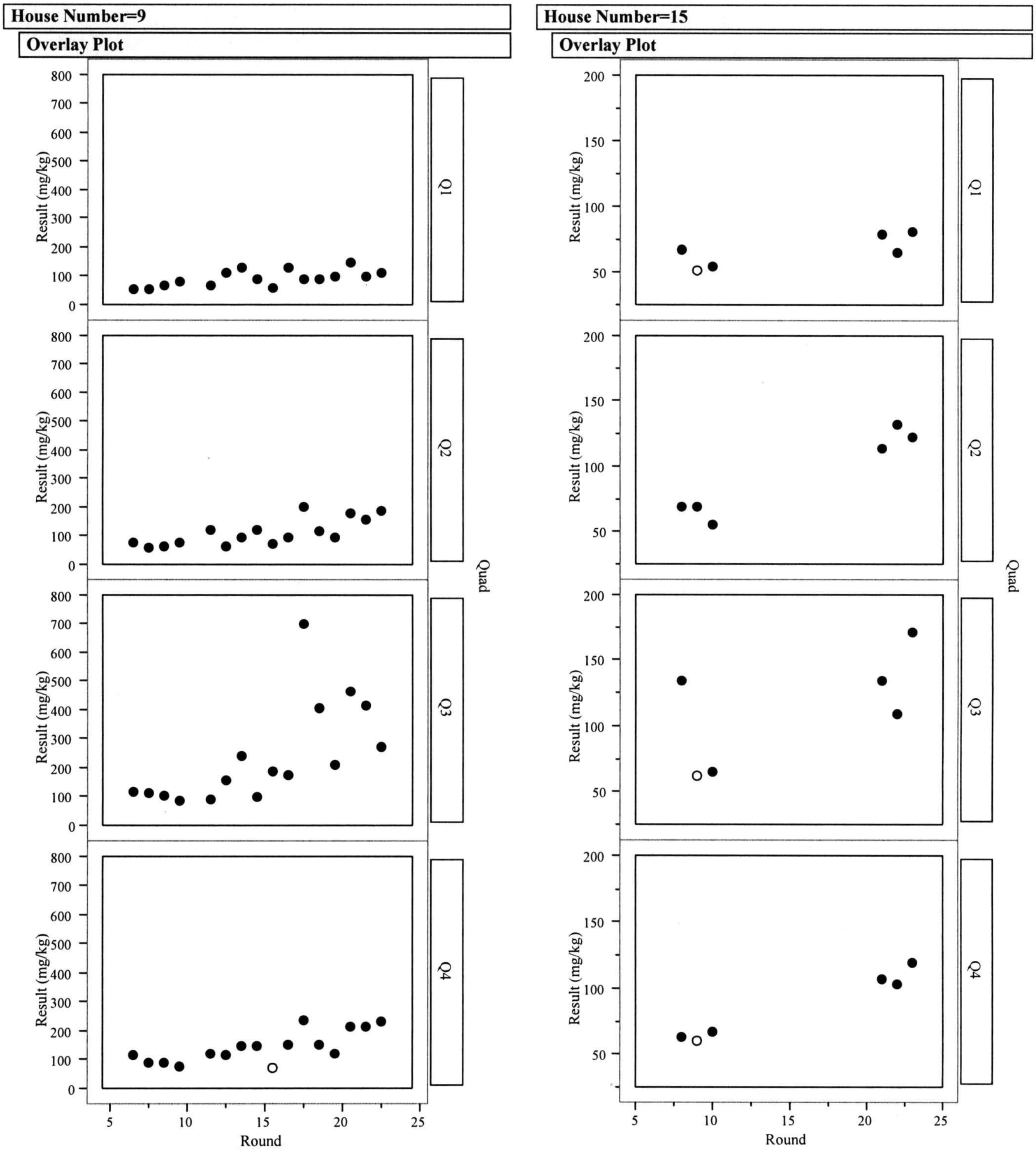


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)

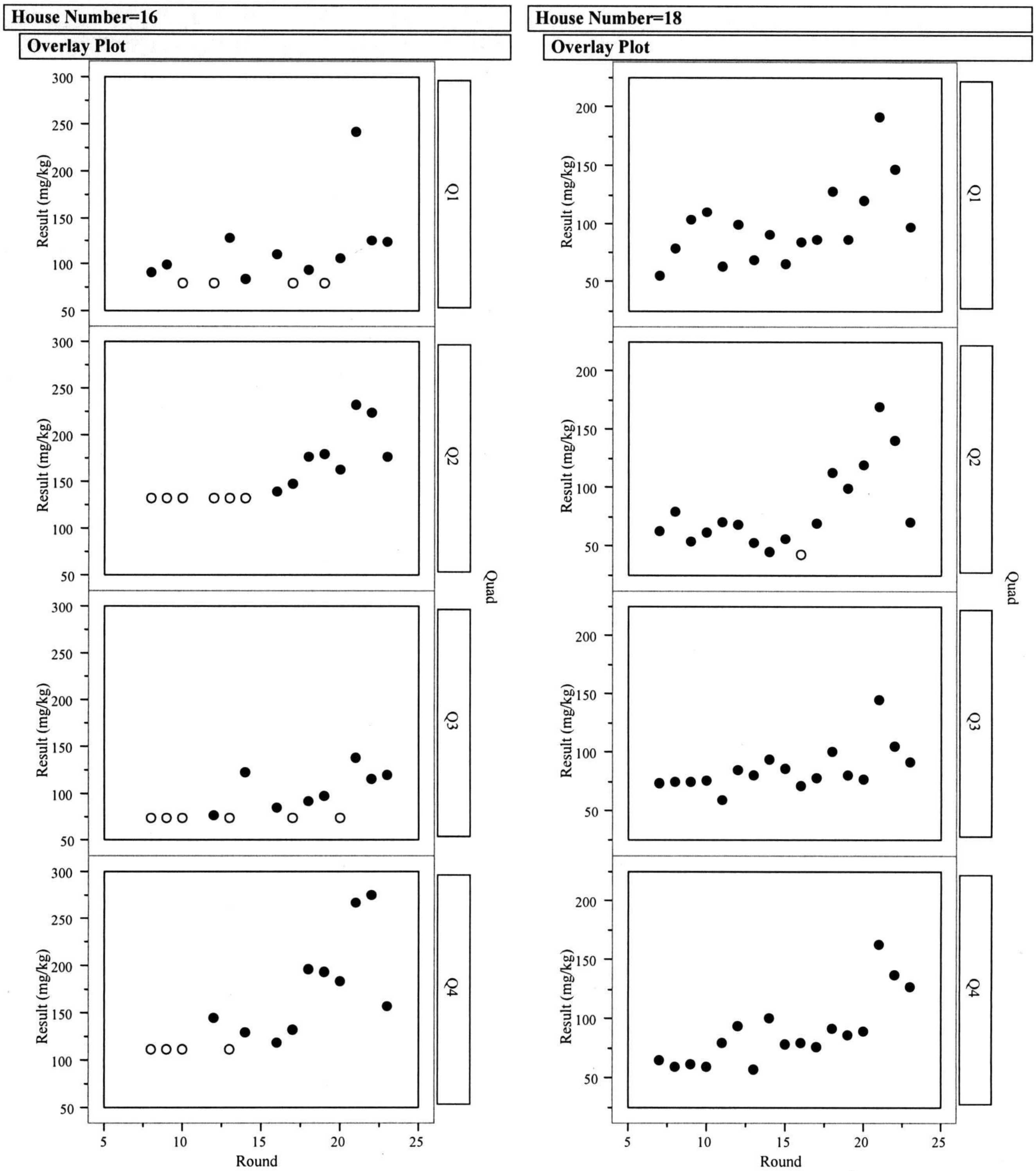
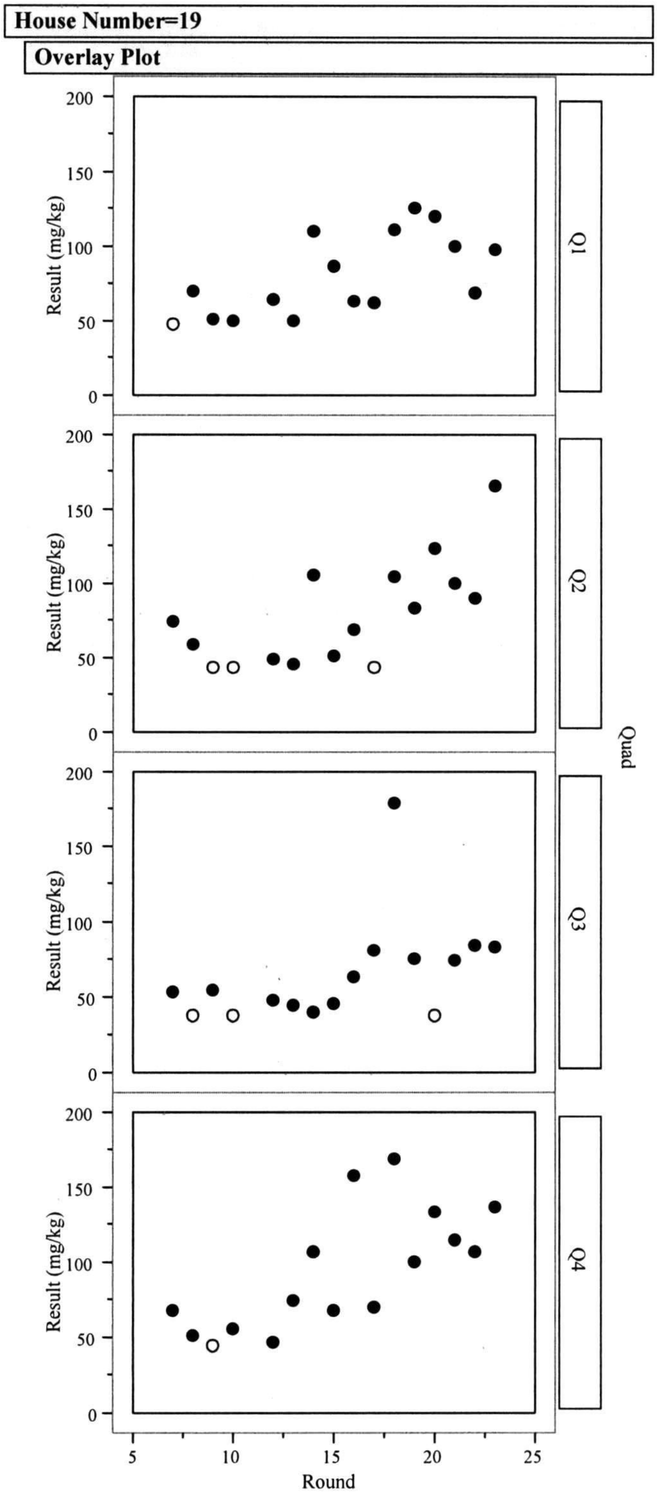


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)



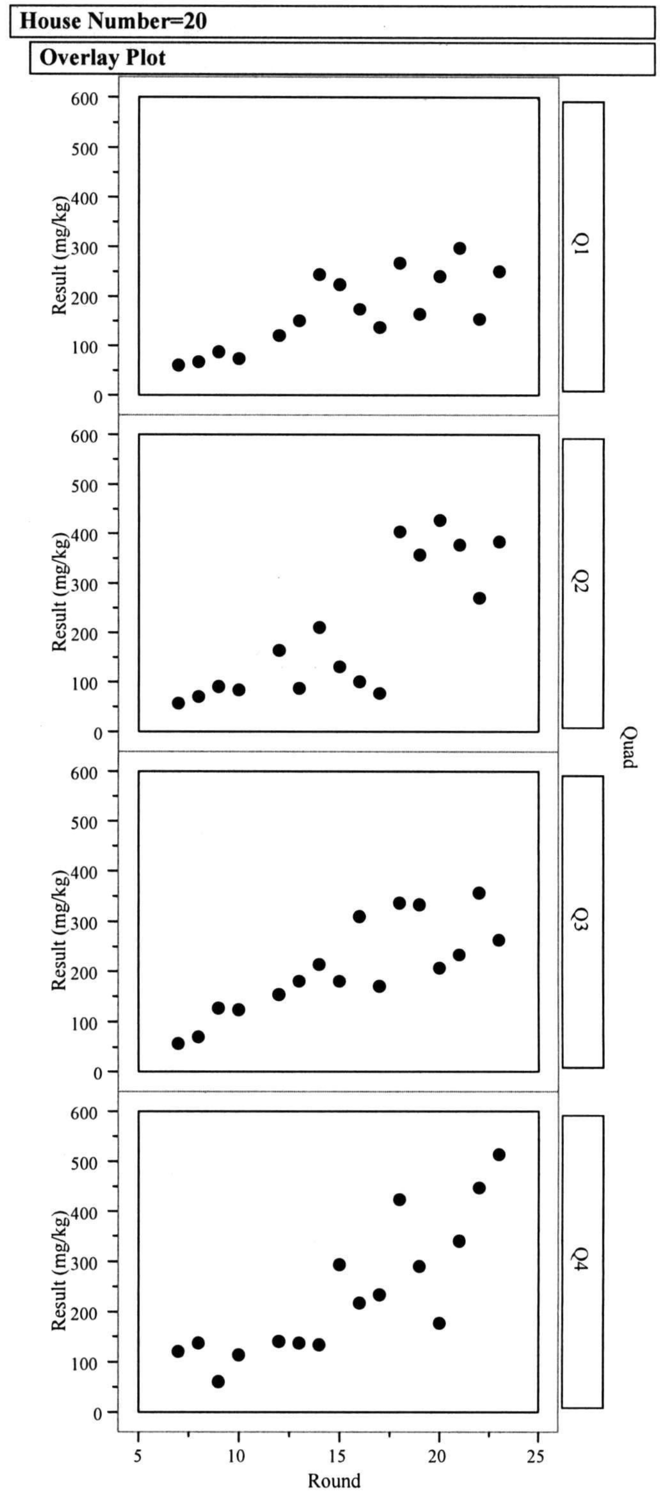
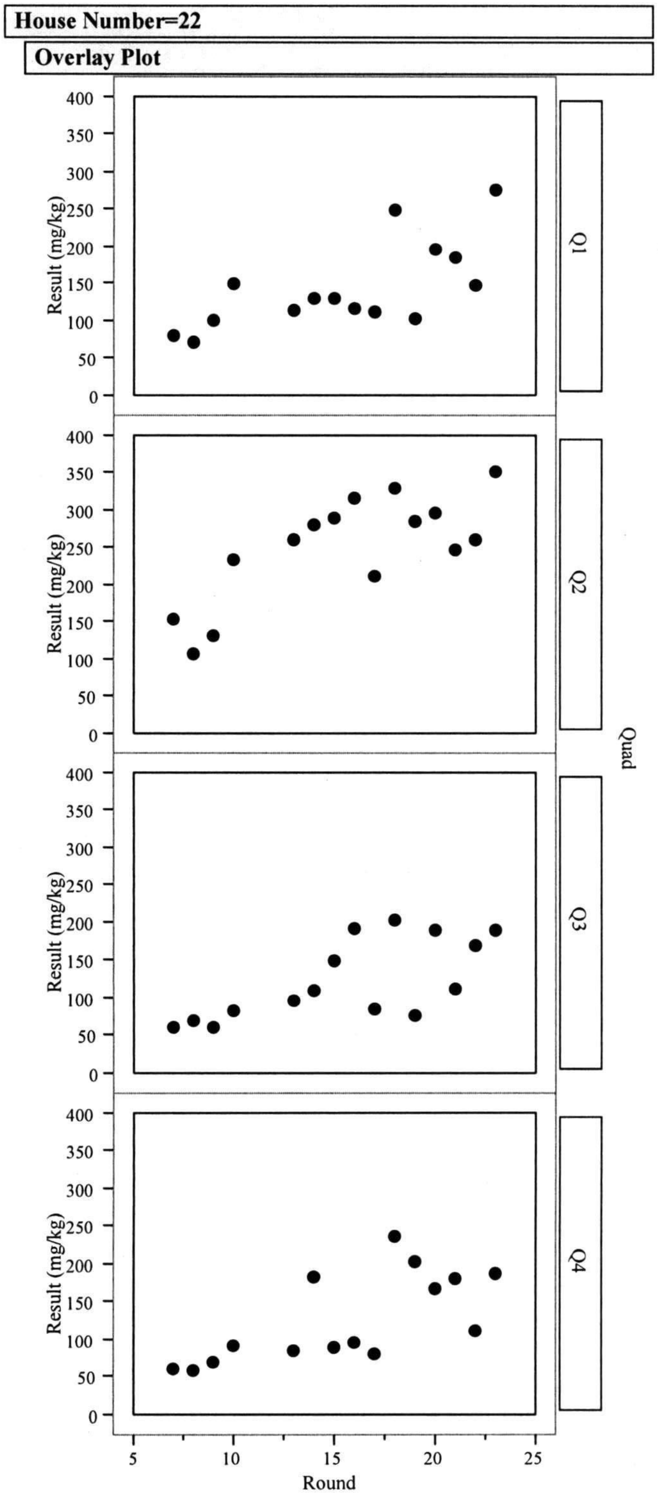


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)



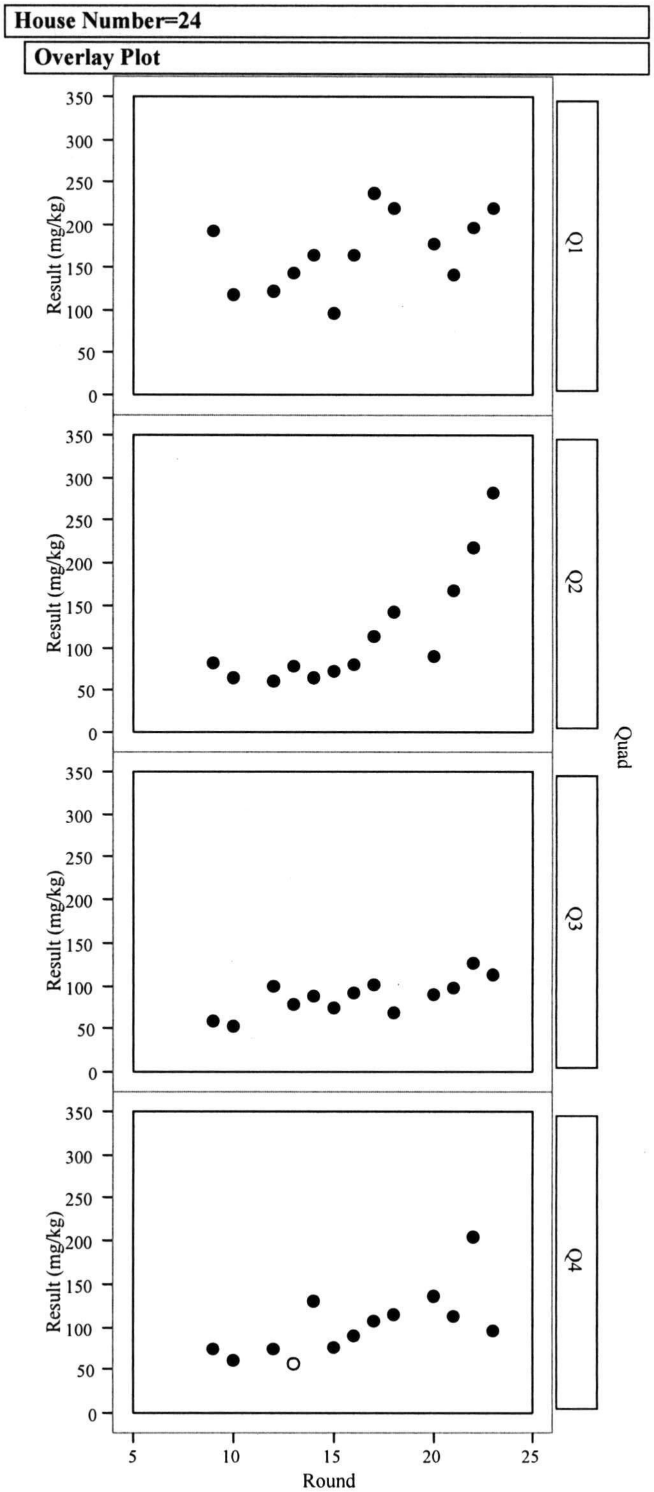
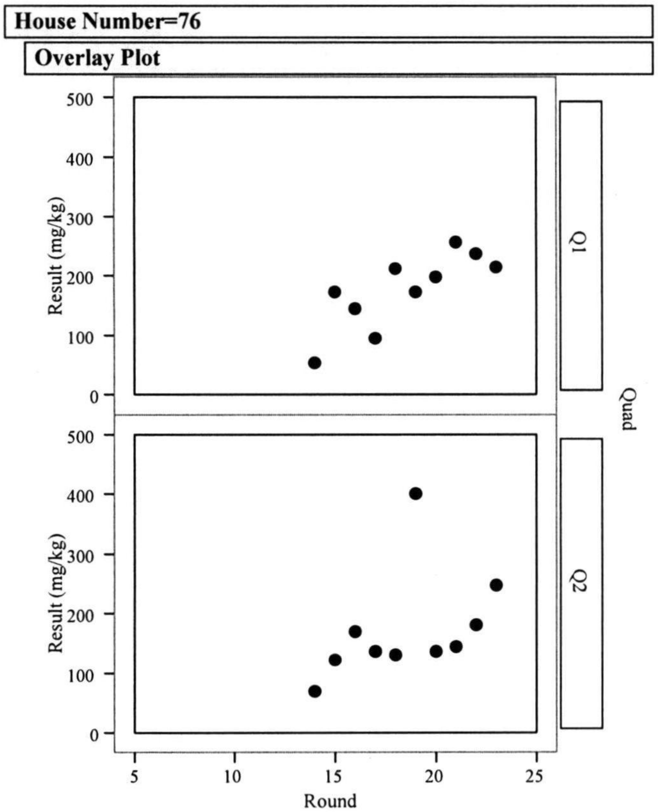


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)



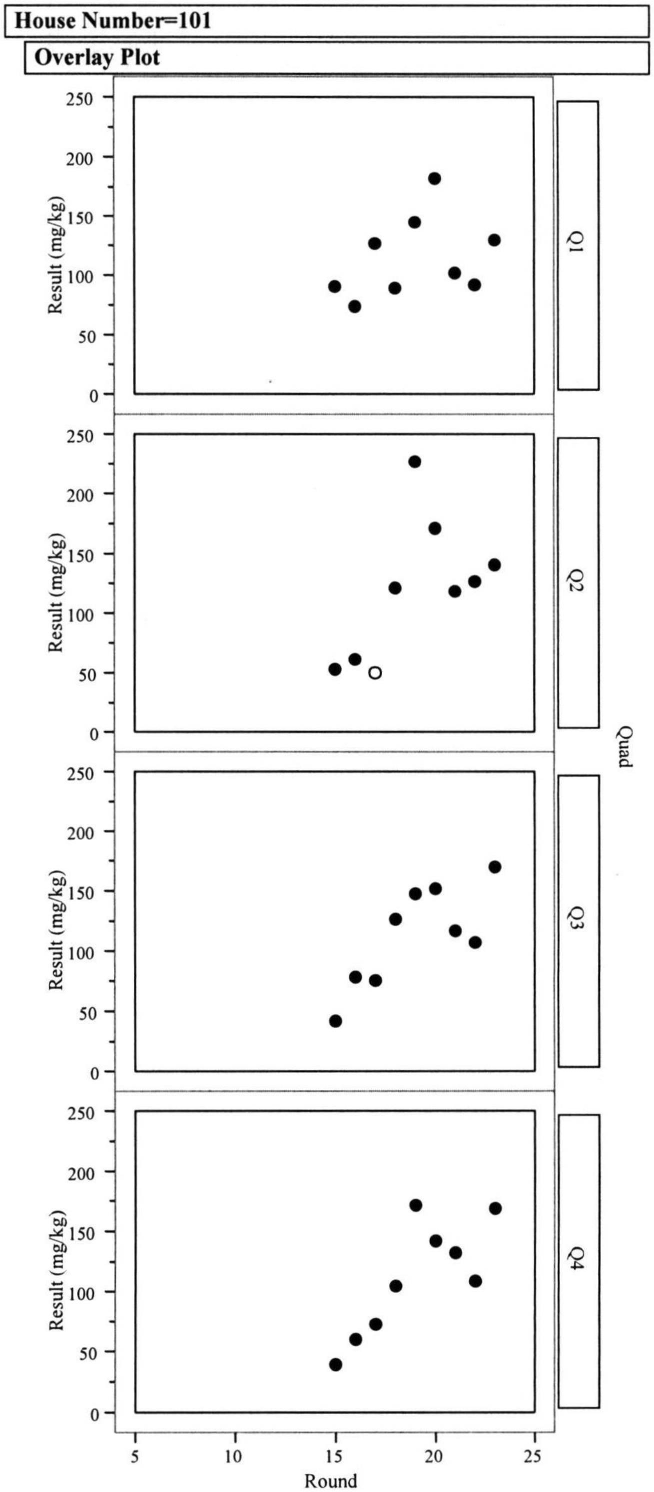
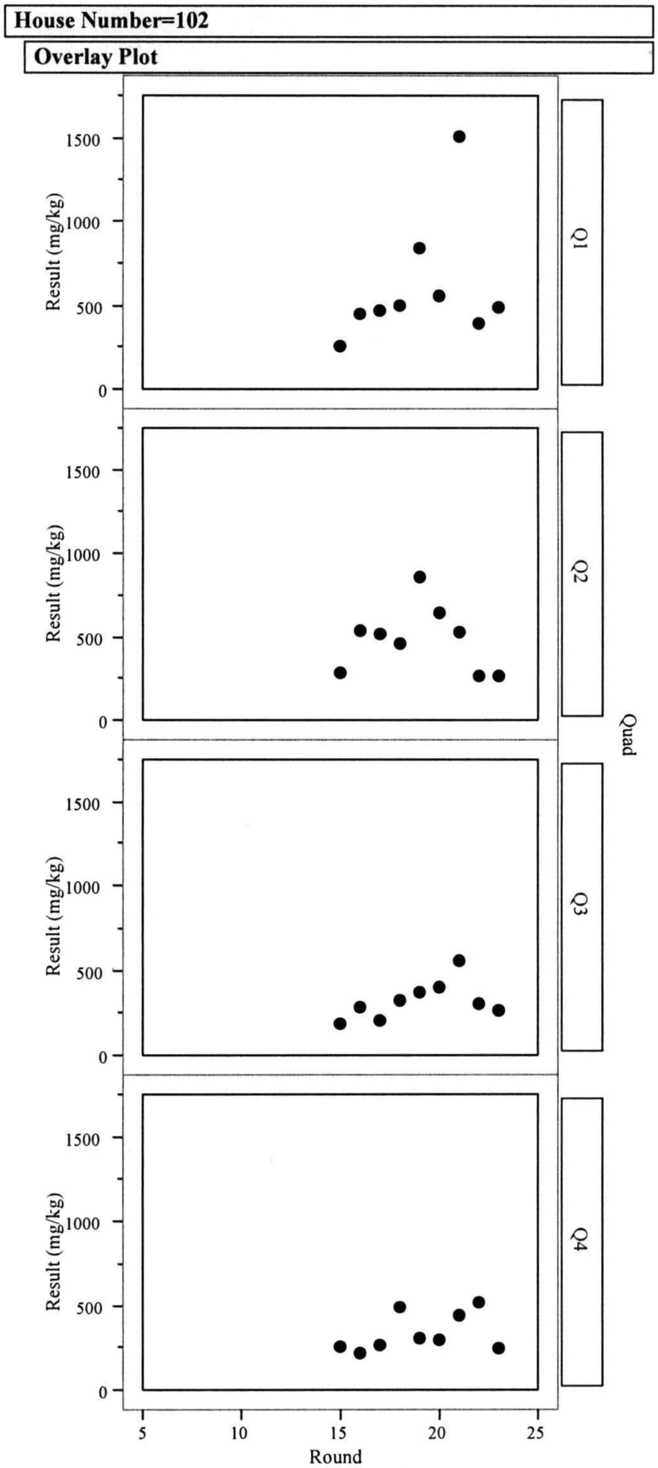


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)



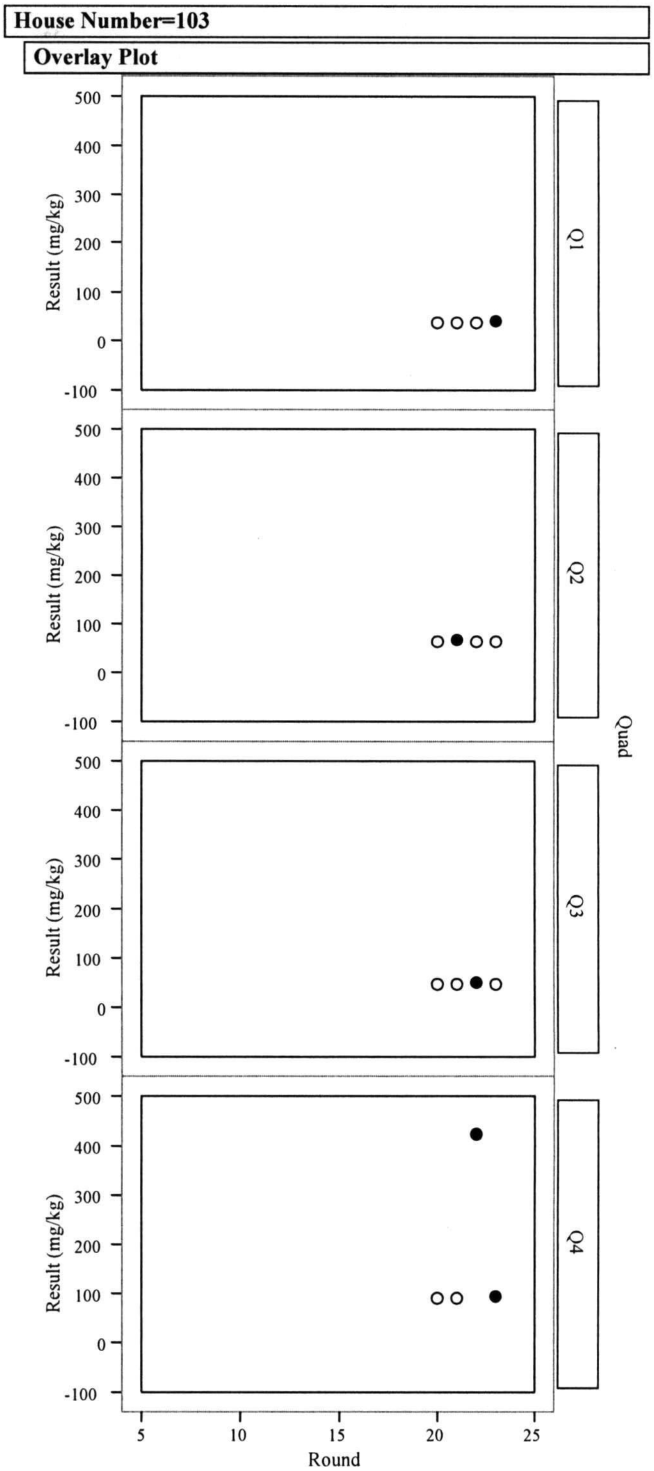


FIGURE 1. Lead Concentration Trends From Round 7 Through 23 (Cont)